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Activity of Personal Protective Gear Worn by Workers Near Fukushima:

Approximating Personal Exposure to Radioactive Cesium In and Near the Fukushima Exclusion

Zone

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Abstract

This study examines the activity of Cesium 137 radiation in air filters and Personal Protective Gear (PPG) worn by remediation workers in and near the Fukushima exclusion zone. Activity is evaluated after radioactive decays are counted using a Broad-Energy Germanium (BEGe) detector, over a constant period of time for each PPG sample. We then use these activity results to calculate the average activity of PPG by region of the body, and display the average Cs-137 activity levels according to the regions of the body with which they are associated. We then approximate the Cs-137 dose to which a remediation worker would be exposed over the course of a year, again, by region of the body, based on the PPG examined in this study. We observe that the highest Cs-137 activity was found in outer-coverings for hands and feet (gloves had an average Cs-137 activity of 498.8 ± 229.4 Bq/g, and footwear had an average Cs-137 activity of 200.4 ± 9.8 Bq/g). Finally, we propose a possible future study.

Introduction

At 5:46 UCT, on March 11, 2011, a 9.0 magnitude earthquake, the epicenter of which lay off of the coast of Sendai, rocked Japan. The earthquake alone, which was the strongest ever recorded in Japan, caused substantial damage in towns across Honshu, especially in Fukushima and Miyagi prefectures (Tanaka, Kimamura, Tanaka, Narabayashi, & Yamamoto, 2015). A Tsunami ensued, striking the Pacific coasts of Tohoku and Kanto (Figure 1). Together, the Great East Japan (Sendai) Earthquake and consequent tsunami lead to roughly 15,000 confirmed fatalities, more than \$200 billion USD in damages, and the displacement of approximately 160,000 residents (Bishop et al., 2011).



Figure 1. Map of Japan showing the location of the Sendai Earthquake epicenter and the nuclear facilities affected (Tharum, 2011).

The inundation, explosions, three meltdowns, and release of radioactive isotopes that occurred at the Fukushima Daiichi nuclear power plant, located in Futaba District, Fukushima Prefecture, resulted from the Great East Japan Earthquake and ensuing tsunami. This plant, located directly on the Pacific coastline, consisted of six nuclear reactors. Following hydrogen explosions at reactors one, three, and four, and severe reactor core damage at unit two, radioactive material, particularly cesium and iodine isotopes, were released into the environment (Hachisuka et al., 2011). This precipitated the establishment of an exclusion zone in Fukushima Prefecture, and the evacuation of all residents from that exclusion zone (Hachisuka et al., 2011). The National Diet, the national bicameral legislature of Japan, formed a commission to investigate the sequence of events that resulted in the explosions at the Fukushima Daiichi power plant, the exact role that the earthquake and resulting tsunami played in the development of this disaster, and the level of responsibility held by Tokyo Electric Power Company (TEPCO) and the national government of Japan (Tanaka, et al. 2015). The report produced by the committee emphasized the need for major institutional reform, but left far more ambiguous

recommendations regarding the health effects of the long-term exposure of residents and cleanup workers to low-dose radiation (Hachisuka et al., 2011). Indeed, the committee remarked that the health effects of long-term exposure to low-dose radioactive cesium was little understood and had not, at the time, been studied extensively by the National Diet (Hachisuka et al., 2011). As a result, the analysis and modeling of health effects (namely, projected mortalities and morbidities) calls for more data reflecting the day-to-day exposure of residents and cleanup workers. Armed with this data, a detailed assessment of low-dose, day-to-day exposure to radioactive isotopes, particularly Cs-137, can be made.

Recent studies have attempted to model and assess the widespread health implications of the Fukushima Daiichi accident, including the work of Ten Hoeve and Jacobson. In their analysis, Ten Hoeve and Jacobson (2012) estimate future mortalities and morbidities by modeling the dissemination of key radioactive isotopes through the atmosphere, and their accumulation in the land and oceans. Their theoretical results are checked against data gathered from Nuclear Test-Ban sensors¹ in Japan and across the Pacific. Beyea et al. (2015) reassess Ten Hoeve & Jacobson's analysis using their own model. These analyses consider aggregate results, while Lee (2012) showed that individuals who have traveled to Japan and who have been exposed to radioactive isotopes from the Daiichi accident are examined.

¹ The Nuclear Test-Ban Sensors that were used to test the model developed by Ten-Hoeve and Jacobson (2012) detect the atmospheric presence of radioisotopes connected with the testing of nuclear weapons.

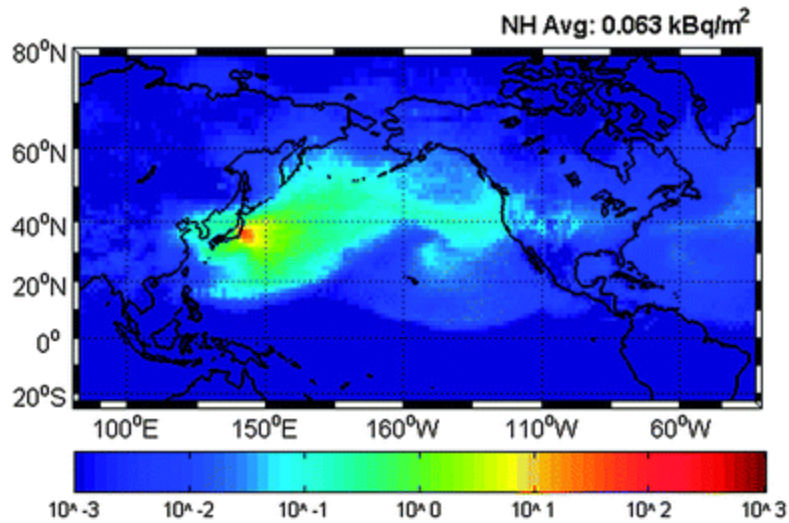


Figure 2. Projected worldwide distribution of Cs-137 following the Fukushima accident (Ten Hoeve & Jacobson, 2012)

The presence of large-scale analyses of aggregate health effects, especially when their models are supported by Nuclear Test Ban sensor data, provide valuable projections, while Lee et al. (2012) provides us with a more individual, yet more narrow, assessment of the health impact. In the study of Lee et al. (2012), the concentrations of dicentric chromosomes² in 265 individuals who had traveled to Fukushima (near, but not within the exclusion zone) were studied, with the ultimate conclusion that travel in Fukushima alone did not produce higher concentrations of dicentricities.

This study use the personal protective gear worn by Fukushima remediation workers, along with samples of clothing, air filters, and dust as our primary sources of data. We use these sources, particularly articles of clothing, to study and model the day-to-day exposure of Fukushima remediation workers and residents, so that new health analyses concerning long-term exposure of individuals to low-dose radioactive isotopes (specifically, Cs-137) may be prepared in the future. We focus on Cs-137 radiation in particular because this radioisotope, along with

² A dicentric chromosome has two centromeres. Lee et al. (2012) considered the concentration of such chromosomes (known to be linked to radiation exposure) to provide a strong method of approximating radiation dose after-the-fact.

Cs-134, is a signature of radiation from nuclear power. Cs-137, however, has a half-life of roughly 30.17 years, while Cs-134 has a half-life of roughly 2.06 years. Cs-137 radiation then remains in higher and more easily detected quantities than Cs-134 radiation.

It should be noted that Cs-137 is also a byproduct of nuclear detonations. Ambient radiation from the testing of nuclear weapons, along with residual Cs-137 radiation from the nuclear detonations at Fukushima and Nagasaki, must then be taken into account. To this end, several blank samples from Japan, along with measurements of our laboratory, were used to establish a background radiation profile for samples from Japan, in the environment in which the study was conducted.³

³ For the ambient radiation profile employed as a background by this study, the reader is directed to the Results section.

Methodology

The objective of this study was to assess the activity of personal protective gear worn by cleanup workers operating within the exclusion zone. To this end, four central research objectives were identified:

1. Select a suitable subset of the provided samples, acquired in the Exclusion Zone in the Fukushima region, such that radiation dose could be mapped onto the surface of the human body.
2. Use a gamma radiation detector to identify and measure the relative presence of important radioactive isotopes in the various samples.
3. Analyze the data collected and relate the presence of radioactive isotopes in the PPG samples to the radiation exposure of both active cleanup workers and the general public.

In the following section, the methods used to achieve the above research objectives are described.

Sample Selection and Organization

Fukushima exclusion zone cleanup workers donated the personal protective gear (PPG) that was investigated in this study. In addition to the PPG, air filter and soil samples were collected from outside of the exclusion zone. In Okinawa, Japan, a pair of work gloves, similar to those worn by the cleanup workers, was purchased. These gloves had not been to Fukushima, and they were used for three “blank” measurements.

Although many samples were provided, 38 non-blank samples were ultimately selected for testing. To provide a survey of the activity associated with clothing worn in various regions of the body (face, arms, hands, torso, waist, legs, ankles, and feet), this study used all available

socks, gloves, Tyvec® sleeves, face masks, and outer footwear. The respirator filter investigated in this study was chosen from among two respirator samples, as the sample most easily mounted to the BEGe detector face (see Sample Testing). Soil and dust samples were chosen to provide an even survey of ground and air radiation, in a variety of locations from inside and outside the exclusion zone. PPG samples were mostly rejected on a basis of size; larger, bulkier samples were difficult to reliably mount on the detector face.

Sample Testing

An unshielded Canberra broad-energy germanium (BEGe) detector (serial number 13010, model #BE2825P), cooled by a constant recycled circulation of liquid nitrogen (Canberra, n.d.), was used to count radioactive decays of various energies. Every sample was fastened to the detector face using Scotch tape.

At least three different samples of each of the following articles of clothing were tested: face masks/respirators, gloves, Tyvec® sleeves, socks, and outer footwear. A reflective vest was also tested, in addition to several different regions of a pair of pants worn by one worker. To accomplish this, we cut the cuffs, pocket flaps, and waistline (including the zipper, belt loops, and button) and placed them in separate bags (Later in this section, the procedure for obtaining subsamples from samples is described). Each subsample was then tested to obtain profiles associated with different regions of the body including the waist, legs, and ankles.

For each sample count, the Canberra BEGe detector counted the number of nuclear decays at various energy levels over a period of 120 minutes. Using Canberra Genie 2000 software installed on a desktop computer linked to the BEGe detector through a Canberra Lynx signal box, the 120-minute counts produced a histogram displaying the number of photon emissions detected at each energy level (Canberra, n.d.).

This study used numerous blank counts, including four 24-hour background counts, and six 120-minute counts of uncontaminated samples from Japan (three counts of the blank gloves purchased in Okinawa, roughly 1,756 kilometers from Fukushima, and three of a blank paper sample obtained in Tokyo, roughly 240 kilometers from Fukushima).

Pants and outer footwear had to be tested in separate components. Before they were cut into subsamples, the pants were massed, and a disposable pair of scissors was obtained. The subsamples described were then produced, placed in Ziploc bags (one bag per subsample), and massed individually. This study used three types of samples to test outer footwear: shoelaces, gaiters (outer coverings attached to a cleanup worker's boots), and dirt from boot soles. Because the boots were received as they were worn by the cleanup worker, the same procedure was followed to obtain subsamples from the work boots: we massed the boots, removed the gaiters and dirt under a fume hood, and then bagged, massed, and labeled the subsamples.

Analysis of Data

Raw data collected was transferred into a Comma-Separated Value (CSV) file format and read into MathWorks MATLAB r2016a. Because the background values were collected over 24 hours, and sample measurements were collected over two hours, the data was first time-normalized, divided by the number of seconds in each sample collection.

A general background profile was produced by averaging the ten blank profiles, intended to represent the average ambient radiation both of the laboratory and of samples originating in Japan. The general background profile was then subtracted from each sample profile. If they existed, substantial differences between the sample counts and the background counts were considered to have originated from the sample.

To obtain the counts per second associated with each sample, the histogram data was converted into a list (from which the general background profile had been subtracted), which was normalized by the length of each count to obtain a count rate.

This study made use of a linear calibration, which established a correspondence between the detection channels of the BEGe detector and the energy levels of electrons emitted from radioactive decay of various isotopes (Figure 3).

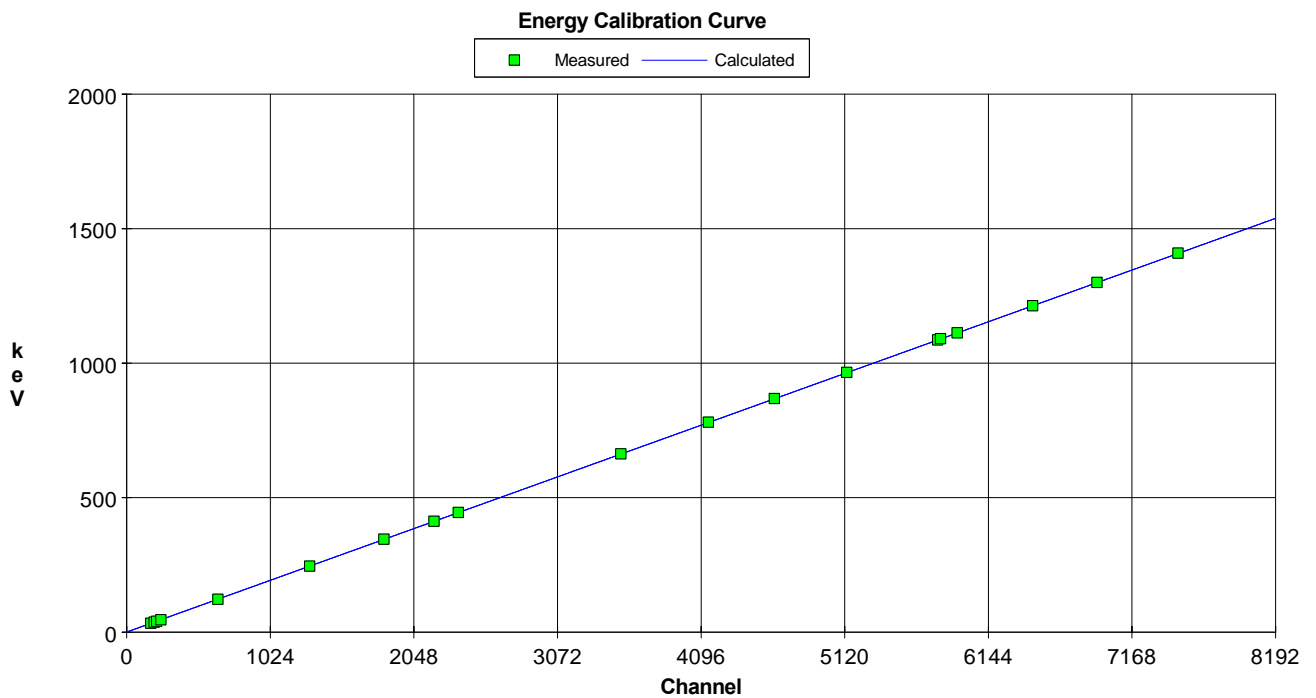


Figure 3. The linear correspondence between electron energy levels and the detection channels of the BEGe detector (Borges, 2017a)

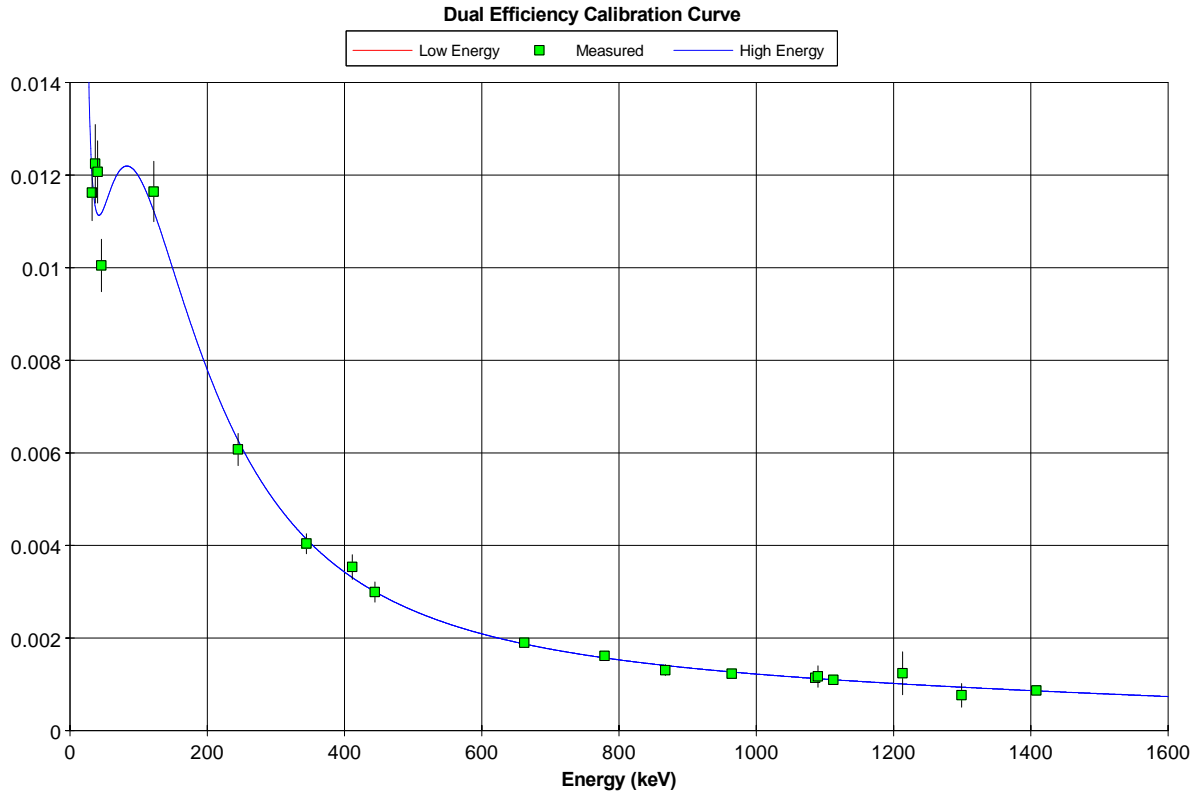


Figure 4. Ratio of radioactive decays detected to radioactive decays, as a function of energy (Borges, 2017b)

A logarithmic curve was provided which was used to calculate the efficiency of the detector (Figure 4). Green points indicate efficiencies calculated using button sources of radioisotopes at the energy levels shown. The blue curve is an approximated efficiency curve calculated by the Genie 2000 software (Canberra, n.d.).

Armed with absolute efficiencies for the energy levels pertinent to Cs-137 and Cs-134, the decays per second associated with each sample were calculated. The mass and count time corresponding to each sample were then used to calculate the activity, in Bq/g.

Results

The data from all samples, shown in net number of decays counted versus energy, as collected is shown in Figure 5.

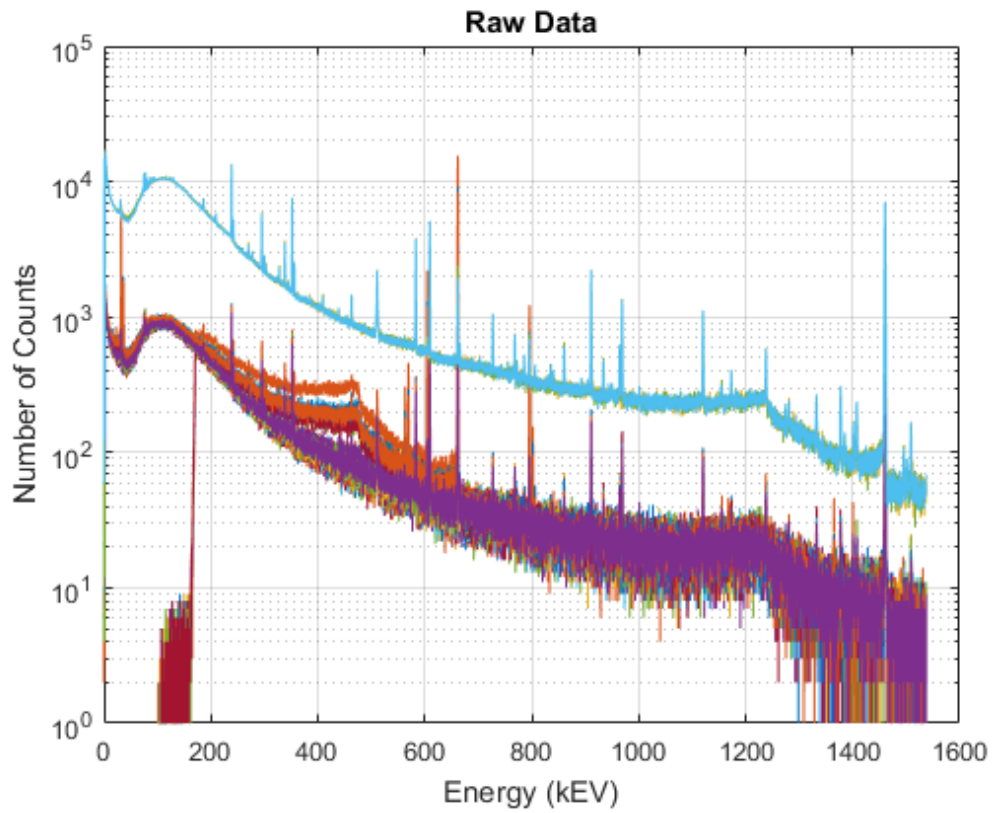


Figure 5. Histogram representation of all raw data collected.

The time-normalized data, presented in counts per second, is displayed in the following profile. (Figure 6).

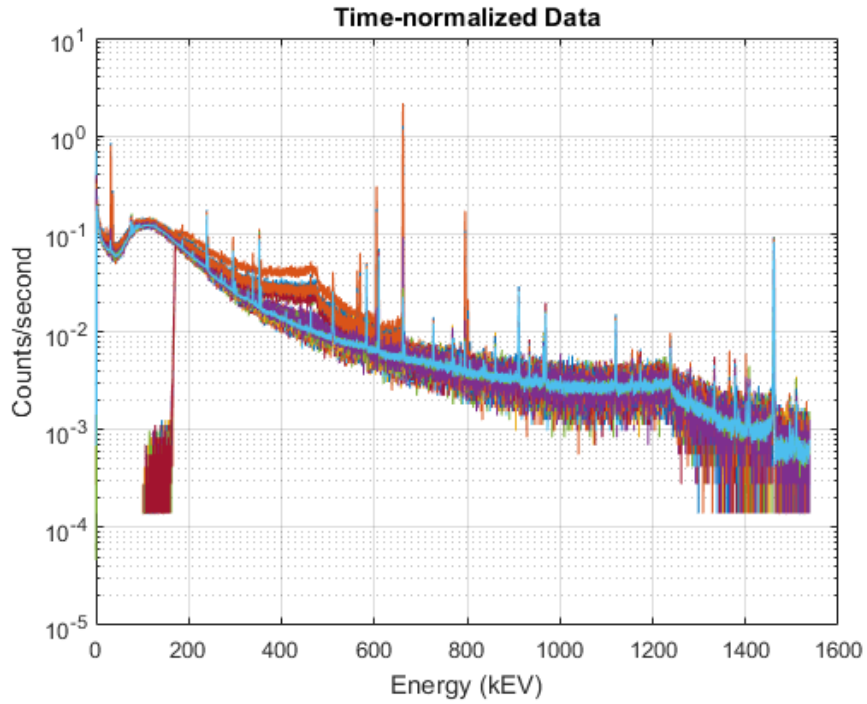


Figure 6. Time-normalized raw data in counts/second

In the following graphic, the average and standard deviations are shown for the background data employed by this study, as described in the methodology (Figure 7).

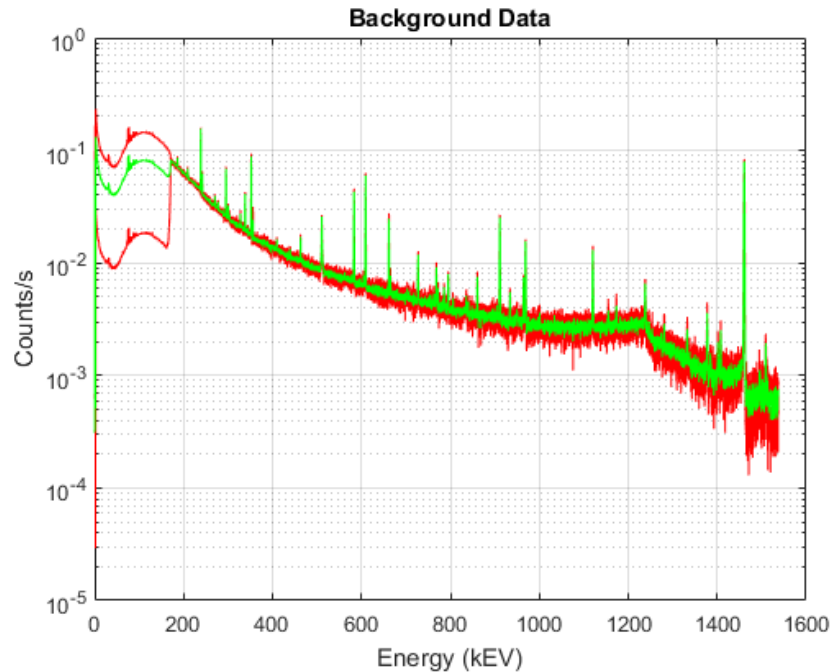


Figure 7. Mean (green) and standard deviations (red) of time-normalized background measurements

In the following series of figures, the process by which this study obtained the average activity of each sample, as described in the methodology, is demonstrated for the same boot sample (one of our footwear samples). Figures 8, 9, 10, and 11 show the progress from net counts, to counts per second over background, to counts per second per gram over background, to Bq/g over background.

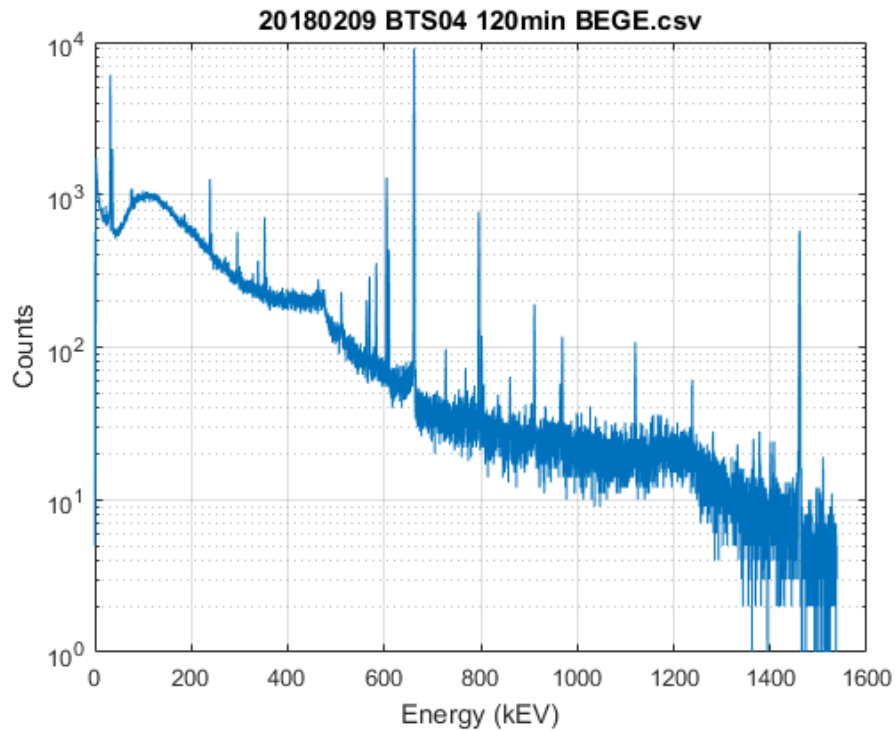


Figure 8. Representative raw count profile for boot sample

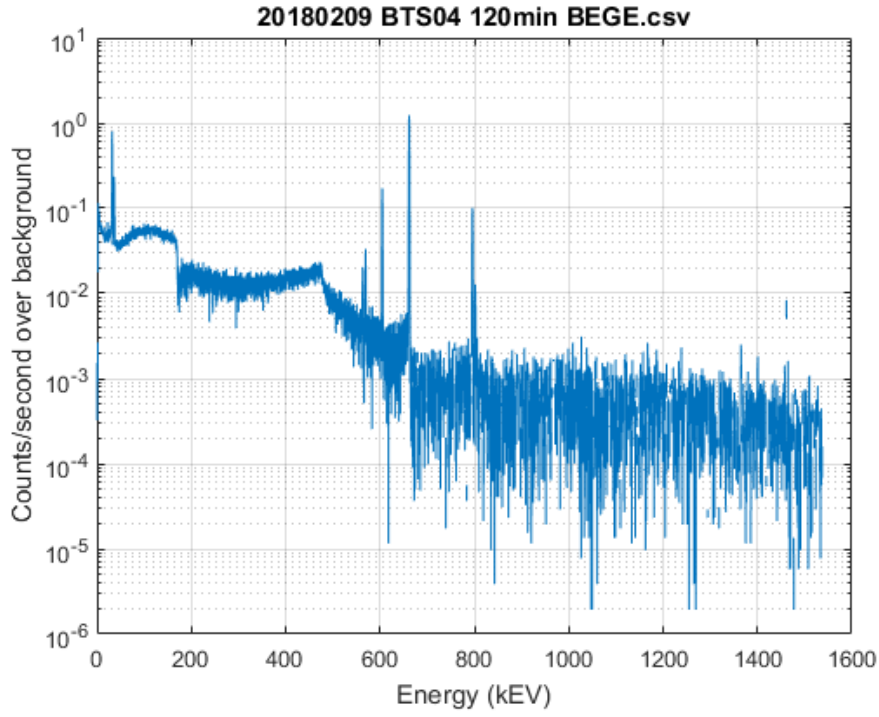


Figure 9. Representative time-normalized count profile of boot sample

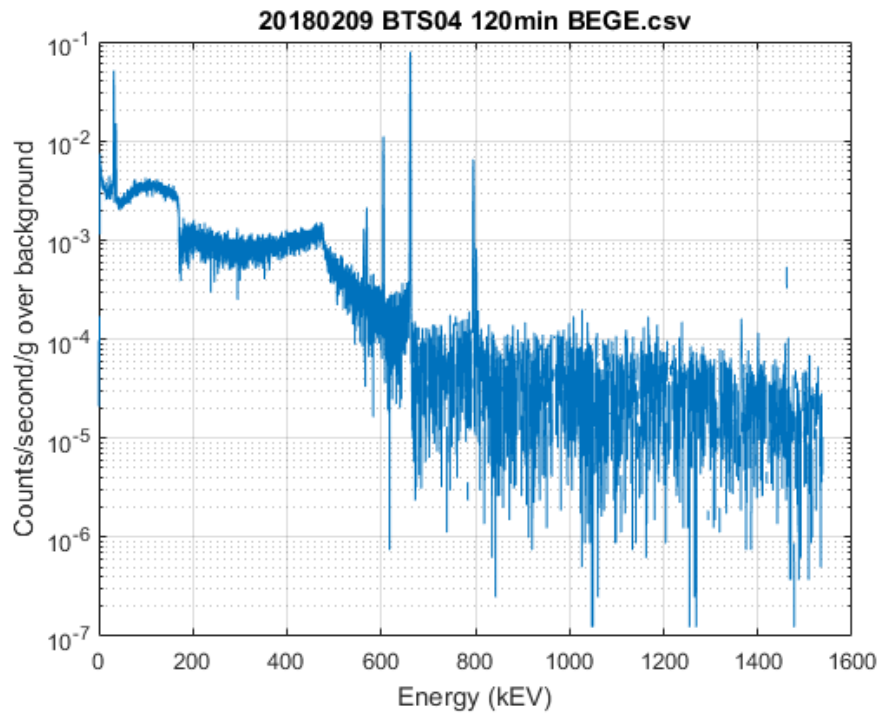


Figure 10. Representative time- and mass-normalized count profile of boot sample.

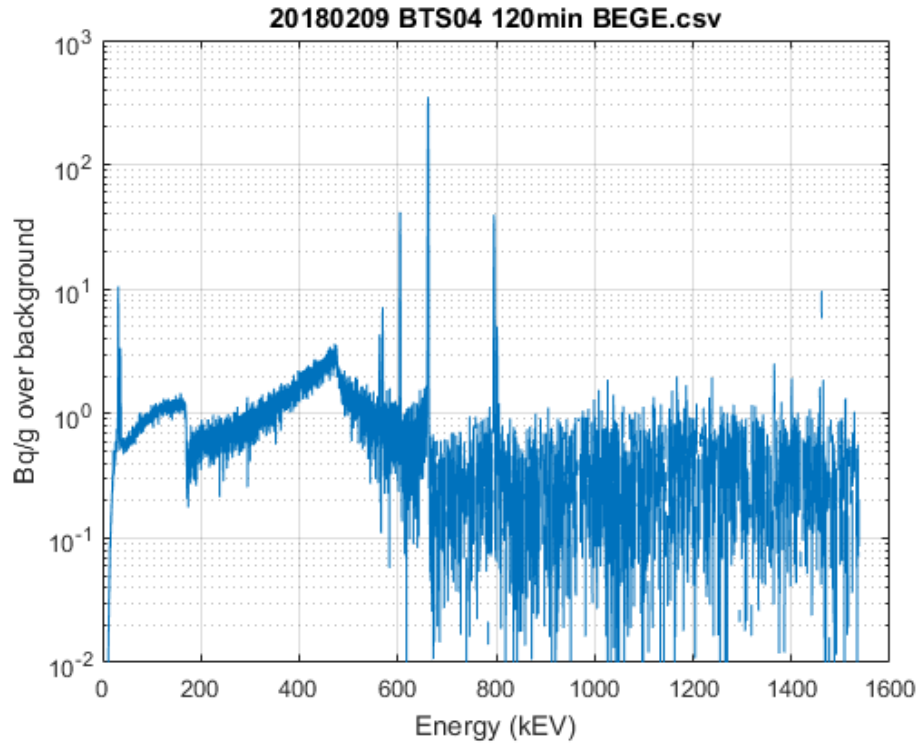


Figure 11. Time-, mass-, and efficiency-normalized count profile from a representative boot sample.

After data collection was complete, the results were aggregated for each category of samples. There were five categories with a sufficient number of samples from which to draw conclusions. These categories consisted of face-covering samples (n=4), such as face masks (Figure 12), hand-covering samples (n=4), consisting of gloves (Figure 13), leg-covering samples (n=7), comprised of subsamples cut from workpants (Figure 14), foot-covering samples (n=10), including boots, socks, and shoelaces (Figure 15), and air samples (n=6), which included all environmental filter samples (Figure 16).

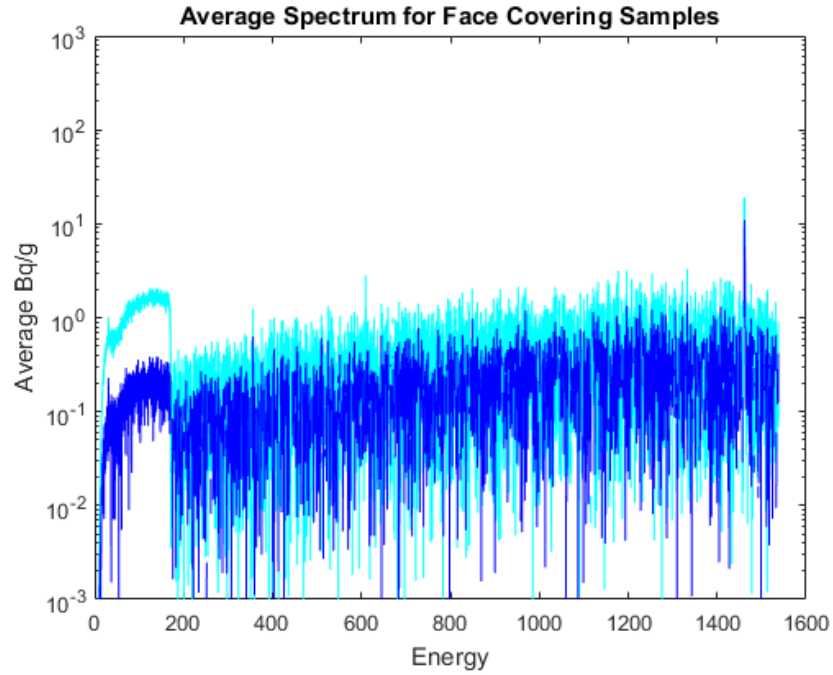


Figure 12. Average activity spectrum (blue) +/- standard deviation (cyan) for face covering samples such as respirators (n=4)

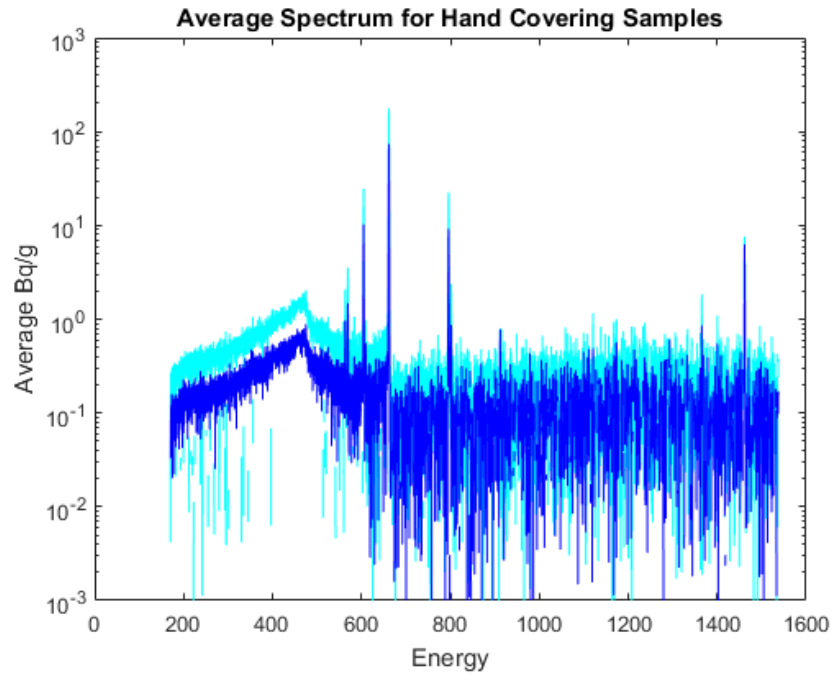


Figure 13. Average activity spectrum (blue) +/- standard deviation (cyan) for gloves (n=4)

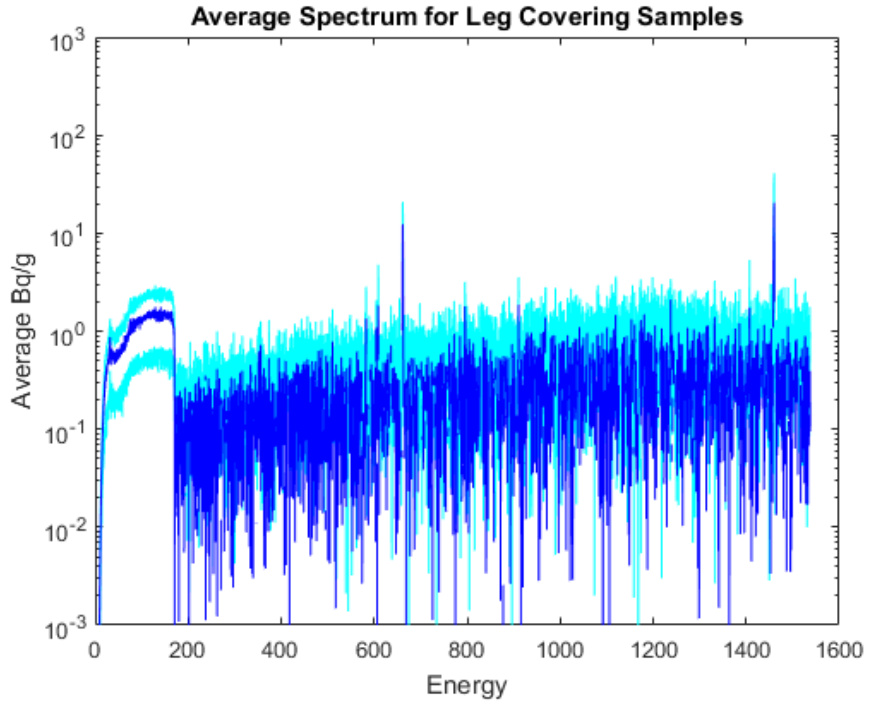


Figure 14. Average activity spectrum (blue) +/- standard deviation (cyan) for subsamples of pants (n=7)

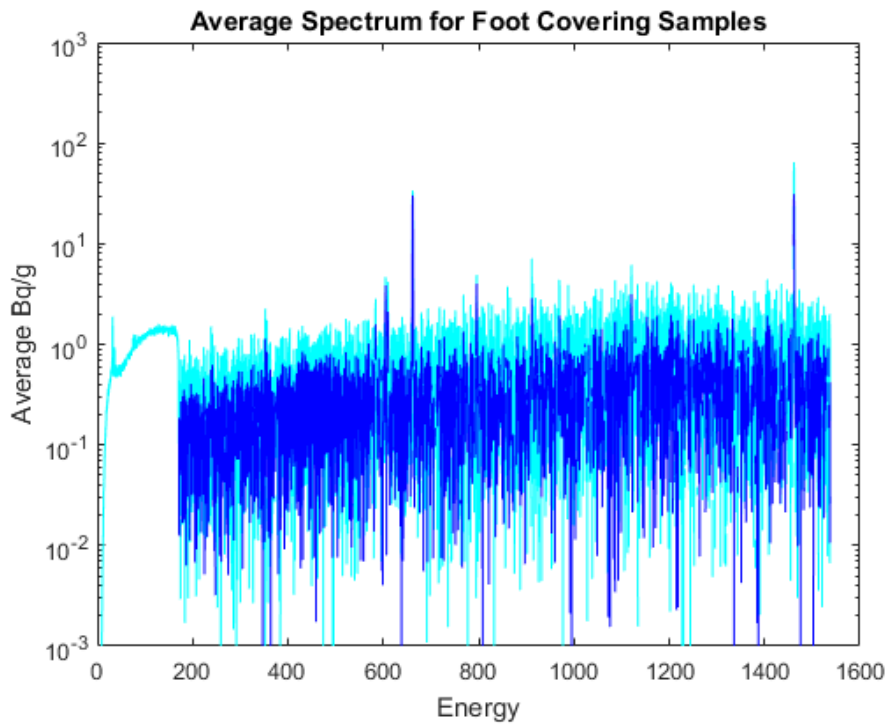


Figure 15. Average activity spectrum (blue) +/- standard deviation (cyan) for foot covering samples such as boots, gaiters, and shoelaces (n=10)

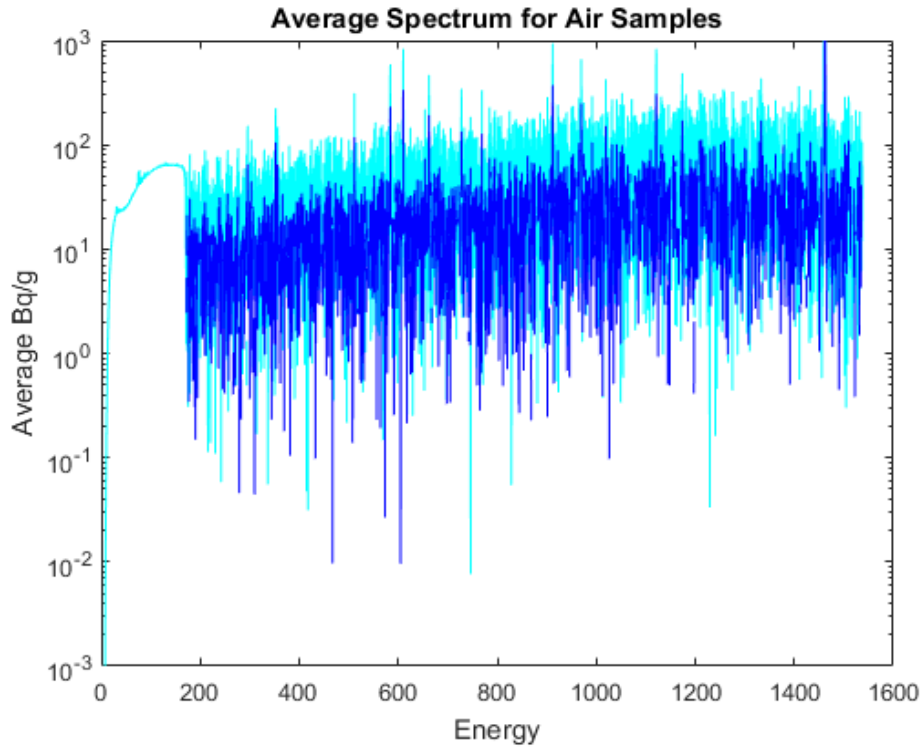


Figure 16. Average spectrum (blue) +/- standard deviation (cyan) for dust-filled samples such as air conditioner filters (n=6)

The sum of the emission counts from 657 keV to 667 keV was taken as the total Cs-137 activity in each category of samples. The sum of averages was taken and the standard deviation was calculated as

$$\sigma_{\Sigma X_i} = \sqrt{\sum \sigma_{X_i}^2}$$

In Figure 17, average Cs-137 activity, as shown by the data, is indicated for samples associated with each investigated region of the body. The table that follows this figure includes the average activity for each region, including standard deviation, given in the following human body illustration.

The rectangle in the air represents the average activity of the air filter samples included in this study.

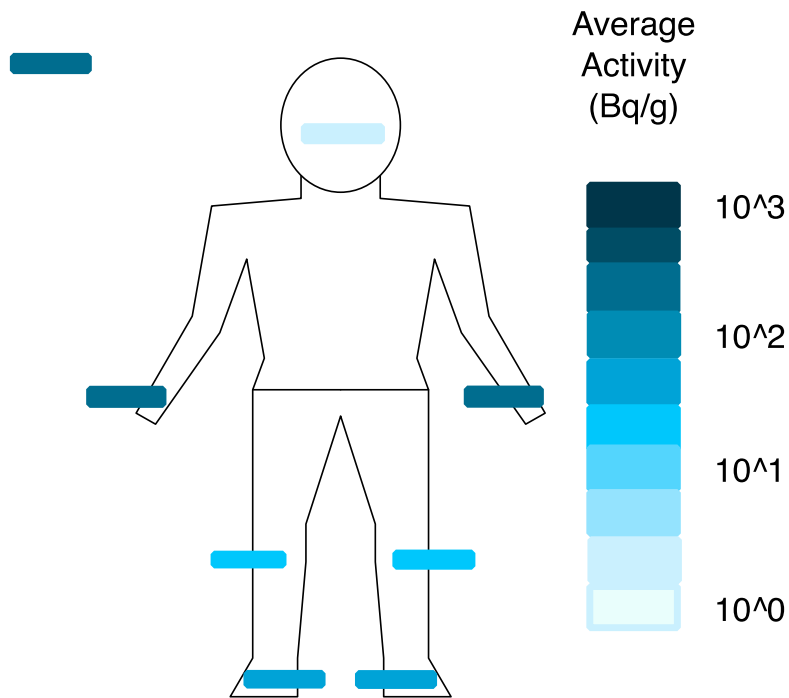


Figure 17. Average activity of PPG associated with different parts of the body; Each rectangle is placed over a measured region. The rectangle on the top-left is representative of the average air filter activities.

Table 1. Average Cs-137 activity by sample category

Region of the Body covered by Sample	Average Activity (Bq/g)
Face	0.0 +/- 2.13
Hands	498.8 +/- 229.4
Legs	72.91 +/- 18.9
Feet	200.4 +/- 9.8
Air	552.5 +/- 408.9

Discussion of Results

Cesium 137

Feet

Foot-coverings also showed significant levels of Cs-137, with average activity of roughly 200.4 Bq/g +/- 9.8. Among the footwear samples, though, there is a range of activity levels. While boot samples showed activity in the magnitude of 10^2 Bq/g, socks showed less activity. In these samples, more Cs-137 activity is found in footwear exposed to outside dirt and air than is found in footwear, such as socks, that were covered by other footwear when worn.

Legs

Leg-covering samples consist of subsamples cut from the workpants. On average, these samples showed Cs-137 activity of roughly 72.9 Bq/g. Although distinct activity was seen at the main Cs-137 peak, the leg-covering samples showed less activity on average, by an order of magnitude, than the foot-covering samples.

Hands

The gloves examined in this study, like the outer footwear, were particularly active, with an average activity of roughly 498.8 Bq/g at the signature energy level for Cs-137 radiation.

Face

The respirators and masks that were tested showed no substantial peak at the signature 662 KeV level for Cs-137.

Many of the samples examined, when worn, were in direct contact with the ground and with dust: outer footwear samples, including the subsamples obtained from work boots, subsamples obtained from trousers, and gloves. Of the 41 samples, gloves and outer footwear comprise nine, and together, they have shown more Cs-137 activity, by a factor of roughly 10 Bq/g, than the samples not exposed to outside dirt and dust. Although the air filters examined in this study showed 552.5 Bq/g of Cs-137 activity on average (with a rather large standard deviation of 408.9 Bq/g), the face masks that were examined showed, on average, no detectable Cs-137 activity.

In the clothing that this study investigated, then, worn by several cleanup workers in and near the Fukushima exclusion zone, Cs-137 exposure was most extreme through direct contact with dirt and dust, and particularly through contact with the ground that leaves sediment.

Cs-137 Dose

This approximates the activity levels of articles of clothing worn by remediation workers in and near the Fukushima exclusion zone over the course of a work-day. We use these calculated Cs-137 activities, combined with the relationship for external dose (Emergency Assistance Center & Site, 2017):

$$D = CA t,$$

Where D is external dosage, C is the surface dose rate in mSv/hBq, specific to each radioisotope, A is Cs-137 activity, and t is time. For Cs-137, $C \approx 8.32 \frac{mSv}{hBq}$ (Waller, Cleary, & Goans, n.d.).

Given the dose relationship above, and the Cs-137 activity levels approximated for the PPG, the following table shows the approximate dose per gram to which a remediation worker would be exposed over an eight-hour workday.

Table 2. Estimated absorbed radiation dose by region of the body

Region	Estimated Dose per Gram (mSv/g)
Hands	$3.3 \times 10^{-2} \pm 1.5 \times 10^{-2}$
Legs	$4.9 \times 10^{-3} \pm 1.3 \times 10^{-3}$
Feet	$1.3 \times 10^{-2} \pm 6.5 \times 10^{-4}$

Limitations

There were several limitations to this study that may have affected the results obtained. While there was a great diversity of samples available for testing, there were more samples available in some categories than others. For instance, only one pair of pants, one reflective vest, and one pair of boots was available for testing. While it was fortunately possible to collect subsamples from these items, samples from other workers in different areas of the exclusion zone and surrounding regions would have improved the accuracy of the results. Additionally, the gamma detector used was not shielded from the surrounding environment, which is a nuclear physics lab where button sources of various radioactive materials are stored. Because these sources were constantly in the room, they were part of the background counts used in the study and so would not constitute a false positive reading. However, because the gamma detector also detected emissions from these sources, the sensitivity to small levels of gamma radiation was reduced and some samples gave a negative value for Cs-137 emission. The data that were

obtained from facemasks, for example, showed Cs-137 activity so low, that negative values were obtained within the standard deviation of 2.13 Bq/g. Such results have no physical meaning, and they are considered to be within the noise of the data obtained.

Conclusion

There is still detectable Cs-137 radiation in the PPG collected for this study. From the calculation above, we find that during a 200-day working year, a radiation worker is likely to receive 2,470 mSv of radiation. This exposure exceeds the U.S. Nuclear Regulatory Commission's annual dose limit for adults in radiation-related occupations of 500 mSv a year (2017). We conclude that remediation workers are primarily exposed to radioactive materials through contact with radioactive soil and dust. The Cs-137 dose of average remediation worker, exposed to Fukushima radiation within the exclusion zone over the course of an entire year, indicates the importance of observing strict radiation limits. Furthermore, the exposure to radioisotopes through exposure to dirt and dust indicates the importance of measures designed to limit the ingestion and inhalation of contaminated material.

By the calculation above, this study approximates that a remediation worker, whose PPG has the average Cs-137 activities calculated, would be exposed to roughly 12.35 mSv of Cs-137 radiation each eight-hour work day. Such a worker would then be able to work only 40 days with Fukushima contamination before the U.S. NRC dose threshold of 500 mSv is reached.

Proposed Future Study

This study examined the contamination of samples, primarily clothing, by radioactive cesium isotopes. Iodine 131, however, with a half-life of 8.02 days, which can cause thyroid cancer by substituting stable iodine in the body, is of more interest than radioactive cesium when

the first two weeks of a nuclear event (i.e., an event in which a substantial amount of radioactive isotopes are accidentally released into the environment) are considered (“TOXNET, ChemIDPlus, Iodine I-131,” n.d.)(Al Eyadeh et al., 2017). The meltdown at Fukushima Daiichi, indeed, released Iodine 131, which can now no longer be measured on Fukushima samples. We therefore propose the following study to examine exposure to radioactive iodine.

To execute the proposed study, uncontaminated articles of clothing would need to be obtained, including a facemask, shirt, gloves, pants, and shoes. The articles of clothing should be placed on a mannequin, 1.85 meters in height, to be placed inside a sealed chamber, consisting of lead walls at least one centimeter in thickness (“TOXNET, ChemIDPlus, Iodine I-131,” n.d.).

A fixed amount of iodine 131 should then be released into the chamber. After a given number of hours, the clothing should be removed from the chamber and divided into subsamples beneath a fume hood. The researchers who remove the clothing from the chamber should wear PPG, including respirators, so that there is no skin exposed to residual iodine. For the purposes of safe disposal, any tools used to cut the clothing into subsamples, along with extra material not saved for testing, should be placed in sealed plastic bags for disposal. The subsamples chosen should provide a survey of various regions of the body, namely the face, neck (collar), arms, torso, waist (including waist pockets), cuffs, and feet. Before subsamples are tested, they should be massed and labeled carefully.

Using a Broad-Energy Germanium (BEGe) detector, energy profiles should be produced for each subsample. The most prominent I-131 gamma energy peak, at 364 keV, should be identified in each profile produced and used to calculate efficiency. We also propose further calculation to approximate the dosage associated with each region of the body represented by the subsamples.

The procedure described in the above paragraphs should be repeated for various amounts of I-131. It should also be repeated with various exposure times, ranging from one hour to 48 hours. The approach described should produce multiple results similar to the results of this study, in which average activity is calculated for clothing samples associated with various regions of the body, and dose is approximated. However, a larger number of trials, with the introduction of controlled variables, should allow for improved statistics.

The relationship between external exposure to radioactive iodine and human thyroid cancer is well known, and as a result of human exposure to iodine radiation through both medical practices and accidental exposure, numerous studies have been conducted to examine iodine radiation (Robbins & Schneider, 2000). In addition, some studies have examined internal, rather than strictly external, human exposure to iodine radiation (Robbins & Schneider, 2000). The proposed study would examine the penetration of clothing, chosen as representative of clothing worn by potential victims of accidental radioiodine exposure, by iodine radiation, approximate dosage, and examine the concentration of iodine radioactivity in various materials and regions of the body. In this way, the proposed study could aid further investigation of the study of accidental human radioiodine exposure.

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